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UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS
AND INTERFERENCES

Ex parte KURT W. KRAMARZ,
RICHARD J. INGRAM, JOHN E. AIKEN,
and GEORGE R. GALLAHER

Appeal 2009-012821
Application 10/716,920
Technology Center 1600

Decided: June 28, 2010

Before RICHARD M. LEBOVITZ, FRANCISCO C. PRATS, and
STEPHEN WALSH, *Administrative Patent Judges*.

PRATS, *Administrative Patent Judge*.

DECISION ON APPEAL

This appeal under 35 U.S.C. § 134 involves claims to methods for the preparation of primary alcohols. The Examiner rejected the claims as obvious.

We have jurisdiction under 35 U.S.C. § 6(b). We affirm.

STATEMENT OF THE CASE

Claims 1, 3-9, and 12-28 are pending and on appeal (App. Br. 4).

Claim 1 is representative and reads as follows:

1. An improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons, the improvement comprising enhancing the selectivity of the crossed-aldol condensation reaction through the use of a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing, wherein the 3-5 carbon aldehyde is propionaldehyde, n-butyraldehyde, isovaleraldehyde, or valeraldehyde, and wherein the water soluble phase transfer catalyst is a quaternary ammonium or phosphonium salt.

The only rejection before us for review is the Examiner's rejection of claims 1, 3-9, and 12-28 under 35 U.S.C. § 103(a) as obvious over Barker et al.¹ in view of Kwok et al.,² Starks et al.,³ Halpern et al.,⁴ or Judge et al.⁵ (Ans. 3-8).

OBVIOUSNESS

¹ U.S. Patent No. 4,426,542 (issued Jan. 17, 1984).

² U.S. Patent No. 5,801,292 (issued Sep. 1, 1998).

³ CHARLES M. STARKS ET AL., PHASE-TRANSFER CATALYSIS, FUNDAMENTALS, APPLICATIONS, AND INDUSTRIAL PERSPECTIVES, 10 and 252-258 (1994).

⁴ Marc Halpern and Reuben Grinstein, *Choosing a Phase-transfer Catalyst to Enhance Reactivity and Catalyst Separation. Part 1*, 236 R. SOC. CHEM. 30-39 (1999).

⁵ GB 1,547,856 (published Jun. 27, 1979).

ISSUE

The Examiner cites Barker as disclosing an aldol condensation reaction performed in the presence of aqueous sodium hydroxide and a water-soluble phase transfer catalyst (Ans. 4). The Examiner concedes, however, that Barker differs from claim 1 with respect to a number of features for which the additional references are cited (*id.* at 5-7).

Specifically, the Examiner finds that Barker does not react mixed aldehydes having different numbers of carbon atoms, and cites Kwok as evidence that the “so-called ‘cross aldol’” reaction was well known in the art, and that it therefore would have been obvious to conduct that reaction using Barker’s methods (*id.* at 5-6).

The Examiner also finds that Barker does not teach removing its phase transfer catalyst by water washing, and cites Halpern as teaching that “water washing has been shown to be an effective means for extracting quaternary ammonium salts from an organic phase” (*id.* at 6). The Examiner also cites Halpern, as well as Starks, as evidence that the three phase reaction system required in certain claims would have been obvious to an ordinary artisan (*id.* at 7-8).

The Examiner cites Judge to show that the requirement in certain claims of using 10 to 50 weight % alkali hydroxide solutions in aldol condensation reactions would have been obvious to a person of ordinary skill (*id.* at 6-7).

Appellants contend that the Examiner’s combination of Kwok with Barker is based on improper hindsight because Kwok teaches that the catalyst used therein, MgO, is critical to the crossed aldol reaction and is disclosed as being water insoluble (App. Br. 11-12). Accordingly,

Appellants urge, Kwok would have directed an ordinary artisan away from using the water-soluble phase transfer catalyst used in Barker and required by the claims (*id.* at 12-13).

Appellants argue that, while Judge performs a cross-aldol reaction, “none of the catalysts listed in the Table at page 4 of Judge et al is considered as ‘water soluble’ by a person skilled in the art” (*id.* at 13). Moreover, Appellants urge, while Halpern provides evidence that water soluble phase transfer catalysts were known in the art, “there is no motivation for a person skilled in the art to combine the use of such water soluble PTC catalyst in an aldol reaction as described by Judge” (*id.* at 14).

Appellants argue that the Specification discloses at least two major improvements with respect to the use of a water-soluble phase transfer catalyst (*id.* at 13 (citing Spec. [0008])). In particular, Appellants urge, in the examples, the Specification shows that a water soluble phase transfer catalyst provides greater selectivity and yield when compared to a water insoluble phase transfer catalyst according to Judge, tricaprylmethylammonium chloride (*id.* at 14).

Given the large commercial scale at which these compounds are produced, Appellants argue, even relatively modest improvements yields provide “significant realized income for the producer. Thus, the yield improvements noted in the Examples of the subject application using a water soluble PTC in accordance with the present invention in comparison to Comparative Example 2 using the catalyst in accordance with Judge et al is quite significant technically” (*id.* at 15).

Appellants have not argued the claims separately. We select claim 1 as representative of the rejected claims. *See* 37 C.F.R. § 41.37(c)(1)(vii).

In view of the positions advanced by Appellants and the Examiner, the issue with respect to this rejection is whether the Examiner has shown that an ordinary artisan would have considered the aqueous based-catalyzed crossed aldol condensation process recited in claim 1, which also uses a water soluble phase transfer catalyst, obvious in view of the cited references.

FINDINGS OF FACT ("FF")

Specification

1. With respect to performing crossed aldol condensation reactions, the Specification discloses:

There are two major improvements resulting from the use of a water-soluble phase-transfer catalyst. The phase-transfer catalyst improves the solubility of the hydroxide catalyst necessary for the crossed-aldol reaction in the longer chain aldehyde, enhancing selectivity for the desired crossed-aldol product versus the self-aldol product produced from the reaction of the shorter chain, more reactive aldehyde. . . . The excellent water solubility of phase-transfer catalysts such as tetrabutylammonium and methyltributylammonium chloride, bromide, or hydroxide derivatives allow for the second major improvement which is the facile recovery of the phase-transfer catalysts from the organic product by aqueous washing.

(Spec. [0008].)

2. The Specification discloses that "[w]ater soluble catalysts which have yielded enhanced selectivity's [sic] equal to or better than the slightly water soluble triacrylmethylammonium chloride PTC are:

hexadecyltrimethylammonium chloride, tetrabutylammonium chloride, tetrabutylammonium bromide, tributylmethylammonium chloride, tetrabutylammonium hydroxide, tetrabutylphosphonium chloride, and

benzyltriethylammonium chloride. Tetramethylammonium chloride gave an unsatisfactory performance” (*id.* at [0018]).

3. Table 1 of the Specification, which appears at paragraph [0019], is reproduced below:

TABLE 1.

Examples of water soluble PTC's for the selective production of 2,4-diethyl-2-octenal (C12) vs 2-ethyl-2-hexenal (C8) from n-butyraldehyde (C4) and 2-ethylhexanal.

Example	PTC	PTC/C4 molar ratio	NaOH/C4 molar ratio	C12 % Yield	C8 % Yield	C12 / C8 molar ratio
Comparative example 1	None	0	1.40	4.7	28.7	0.2
Comparative example 2	Tricaprylmethyl- ammonium chloride	0.09	1.44	42.3	24.6	1.7
Comparative example 3	Tetramethylammonium chloride	0.21	1.34	17.0	33.9	0.5
Example 1	Hexadecyltrimethyl- ammonium chloride	0.08	1.36	47.4	18.1	2.6
Example 2	Tetrabutylammonium chloride	0.09	1.36	57.5	13.1	4.4
Example 3	Tetrabutylammonium bromide	0.10	1.29	58.6	15.4	3.8
Example 4	Tributylmethyl- ammonium chloride	0.07	1.26	61.1	9.6	6.4
Example 5	Tetrabutylammonium hydroxide	0.09	1.34	61.7	11.2	5.5
Example 6	Tributylmethyl- ammonium chloride	0.04	1.23	51.8	21.2	2.4
Example 7	Tributylmethyl- ammonium chloride	0.02	1.44	38.8	32.5	1.2
Example 8	Tributylmethyl- ammonium chloride	0.09	0.69	41.3	30.1	1.4
Example 9	Tributylmethyl- ammonium chloride	0.09	0.35	20.0	57.4	0.3
Example 10	Tributylmethyl- ammonium chloride	0.09	2.13	52.6	7.6	6.9
Example 11	Tetrabutylammonium hydroxide	0.09	0.00	52.7	22.2	2.4
Example 12	Tetrabutylphosphonium hydroxide	0.10	0.00	40.0	40.6	1.0
Example 13	Benzyltriethyl- ammonium chloride	0.10	1.31	41.4	27.6	1.5

The table shows a comparison between the products and yields produced according to the processes used in Comparative Examples 1 to 3,

and Examples 1 to 13. The Table lists the catalysts used and the relative yields of the more desired (C12) and less desired (C8) aldehydes.

4. Based on the data presented in Table 1, the Specification concludes that the “preferred quaternary cations for the aldol reaction are tetrabutylammonium and tributylmethylammonium” (*id.* at [0020]).

Barker

5. Barker discloses “a process in which mixed butenes are converted to a ten-carbon plasticizer alcohol” (Barker, col. 1, ll. 60-62).

6. Barker’s process “utilizes an oxo reaction of olefins, followed by an aldol condensation” (*id.* at col. 2, ll. 52-53).

7. Barker discloses that the “aldol reaction can utilize strongly alkaline catalyst, such as sodium and potassium hydroxide, or sodium and potassium cyanide” (*id.* at col. 3, ll. 51-53).

8. Barker discloses that alcohols having more than ten carbon atoms, useful in preparing detergents, can also be prepared by the disclosed methods (*see id.* at col. 5, l. 5 through col. 6, l. 43).

9. Example 14 of Barker, cited by the Examiner, describes the preparation of such “detergent range alcohols” (*id.* at col. 18, l. 50).

10. In Example 14, the aldol condensation reaction was performed by adding hexanal to 2-methylpentanal to 2-ethylbutanal (*id.* at col. 18, ll. 19-41). Specifically, the “branched aldehydes were placed in a flask with 110 ml of 0.8 M NaOH, and tetrabutylammonium chloride in an amount molecularly equivalent to the NaOH. The tetrabutyl-ammonium chloride serves as a phase transfer catalyst. The reaction flask was heated to reflux and addition [of hexanal] was started” (*id.* at col. 18, ll. 22-27).

11. Barker discloses that “[t]he phase transfer catalyst was effective in improving conversion in this procedure, but use of co-solvents, such as methanol or diols, may be more practical for large scale continuous operations” (*id.* at col. 18, ll. 40-43).

Kwok

12. Kwok is directed to “to a process for condensation of aldehydes, and more particularly to a process for forming desired aldols by the reaction of specified aldehydes in the presence of a novel catalytic agent” (Kwok, col. 1, ll. 6-9).

13. Kwok discloses that the “reaction of aldehydes in the so-called ‘aldol condensation’ reaction is well known” (*id.* at col. 1, ll. 14-15).

14. Kwok also discloses:

As is also commonly known, the aldol condensation reaction may be used not only for the condensation of a given aldehyde, but for the combination of different aldehydes, producing a so-called “cross aldol”, provided that at least one of the aldehydes contains an α hydrogen. For example, isobutyraldehyde may be reacted or condensed with formaldehyde in the presence of alkali, under suitable conditions, to form hydroxypivaldehyde. The hydroxypivaldehyde may, if desired, be hydrogenated to form neopentyl glycol, a valuable commodity chemical.

(*Id.* at col. 1, ll. 58-67.)

15. Kwok discloses that both the aldol condensation reaction and the cross aldol reaction can be performed “with a catalytic amount of hydrated MgO” (*id.* at col. 2, ll. 36-66).

16. Specifically, Kwok advises:

A critical aspect of the invention is the use of hydrated MgO for catalyzing or promoting the condensation reaction. As understood herein, the phrase “hydrated MgO” refers to solid particulate MgO (magnesium oxide) which has been treated or contacted with sufficient water (including wet steam) for a sufficient period of time (temperature and pressure are not significant except to the extent required to have water present) to effect certain changes in surface morphology of the solid particles. Normally, hydration of the MgO may be carried out simply by mixing the particulate MgO with ample water and allowing the mixture to stand at room temperature, e.g., 20° C., for a sufficient period of time, e.g., 15 or more hours.

(*Id.* at col. 4, l. 58, through col. 5, l. 3.)

Halpern

17. The Halpern publication is a “report . . . directed at providing useful information to process chemists and engineers for choosing commercially available phase-transfer catalysts to enhance reactivity and separation” (Halpern 30).

18. Halpern discloses that “[q]uaternary ammonium, ‘quat’, salts are by far the most common phase-transfer catalysts used in industrial applications. Three commercially available quats will be described in this presentation: Aliquat® 336 (methyl tricapryl ammonium chloride . . .), Aliquat® 100 (tetrabutyl ammonium bromide) and Aliquat® 175 (methyl tributyl ammonium chloride)” (*id.*).

19. Halpern conducted experiments to determine the degree to which those phase transfer catalysts were partitioned into the aqueous phase of an aqueous/organic biphasic milieu to determine the ease by which the catalyst could be removed from the organic product (*see id.* at 31-34).

20. Based on its experiments, Halpern found that, under its test conditions

[O]ne could perform only *half of the water wash* (reducing aqueous waste stream volume) and still *achieve more than an order of magnitude less residual quat salt* using Aliquat[®]175 compared to using Aliquat[®]100. Using two fresh water washes in this [test] system, the residual level of Aliquat[®]175 may be four orders of magnitude less than for Aliquat[®]100 and would be undetectable by commonly known analytical methods.

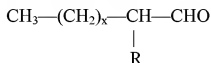
(*Id.* at 35.)

Judge

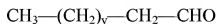
21. Judge discloses an “aldol condensation . . . effected in a reaction medium comprising an aqueous alkali metal hydroxide solution and a phase transfer catalyst, hereinafter abbreviated to PTC” (Judge 2:11-13).

22. Judge discloses that “use of PTC leads to a much greater yield and much greater conversion of starting material than when the aldol condensation is effected in the absence of PTC” (*id.* at 2:14-16).

23. Judge discloses that its aldol condensation reactions can be performed with a “first aldehyde [which] has the formula”



and with a “second aldehyde [which] has the formula”



in which x and y can be 0, 1, 2, 3, or 4, and in which R is “as required for the unsaturated aldehyde product” (*id.* at 2:3-10).

24. Appellants concede that “Judge et al discloses a process using a PTC in a cross-aldol reaction” (App. Br. 13).

25. At page 4, Judge discloses a list of about 46 compounds that “can be used with excellent results” (Judge 4:1) in its aldol condensations (“Phase Transfer Catalysts (a)” (*id.* at 4:5-51).

26. Appellants contend, and the Examiner does not dispute, that none of the compounds listed at page 4 of Judge would be considered “water soluble” by an ordinary artisan (App. Br. 13).

27. At page 5, Judge discloses additional “PTCs which can be used with excellent results in the process of this invention” (Judge 5:6).

28. Judge discloses that these additional PTCs include but are not limited to those having the formula



in which:

(a) X^{z-} is an anion (e.g. sulfate, phosphate, acetate, nitrate, chloride or bromide);

(b) $z-$ is an integer denoting the number of negative charges on each anion;

(c) y is an integer equal to z , so that the cation and anion charges balance;

(d) R_1 , R_2 , R_3 , and R_4 are separately selected from (i) alkyl and alkenyl groups having 1 to 20 carbon atoms and (ii) aralkyl groups having 7 to 16, preferably 7 to 12 carbon atoms, e.g. benzyl;

(e) A is N, P, or As. It is generally preferred that A be N or P.

Preferred PTCs include tricaprylmethylammonium chloride and didecyldimethylammonium chloride.

(*Id.* at 5:7-17.)

PRINCIPLES OF LAW

In *KSR Int'l Co. v. Teleflex Inc.*, 550 U.S. 398 (2007), the Supreme Court reaffirmed that “when a patent ‘simply arranges old elements with each performing the same function it had been known to perform’ and yields no more than one would expect from such an arrangement, the combination is obvious.” *Id.* at 417 (quoting *Sakraidia v. Ag Pro, Inc.*, 425 U.S. 273, 282 (1976)).

Regarding the obviousness question, the Federal Circuit has advised that “a finding that the prior art as a whole suggests the desirability of a particular combination need not be supported by a finding that the prior art suggests that the combination claimed by the patent applicant is the preferred, or most desirable, combination.” *In re Fulton*, 391 F.3d 1195, 1200 (Fed. Cir. 2004).

Moreover, “[m]ere improvement in properties does not always suffice to show unexpected results. . . . [W]hen an applicant demonstrates *substantially* improved results . . . and *states* that the results were *unexpected*, this should suffice to establish unexpected results *in the absence of* evidence to the contrary.” *In re Soni*, 54 F.3d 746, 751 (Fed. Cir. 1995).

Also, to establish that claimed subject matter yields an unexpected result, the claimed subject matter must be compared to the closest prior art. *In re Baxter Travenol Labs.*, 952 F.2d 388, 392 (Fed. Cir. 1991) (“[W]hen unexpected results are used as evidence of nonobviousness, the results must be shown to be unexpected compared with the closest prior art.”).

ANALYSIS

We are not persuaded that the Examiner failed to show that an ordinary artisan would have considered the aqueous based-catalyzed crossed aldol condensation process recited in claim 1, which also uses a water soluble phase transfer catalyst, obvious in view of the cited references.

Claim 1 recites an improved process for the preparation of a primary alcohol by hydrogenating an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons. Claim 1 requires the 3-5 carbon aldehyde to be propionaldehyde, n-butyraldehyde, isovaleraldehyde, or valeraldehyde.

Claim 1 specifies that the improvement comprises enhancing the selectivity of the crossed-aldol condensation reaction by using “a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing.” Claim 1 further specifies that the water soluble phase transfer catalyst must be a quaternary ammonium or phosphonium salt.

As disclosed by Kwok, and apparently conceded by Appellants’ use of the Jepson improvement-type claim format, the crossed-aldol condensation reaction between aldehydes with different numbers of carbon atoms was well known in the art (FF 14). *See also Sjolund v. Musland*, 847 F.2d 1573, 1577 (Fed. Cir. 1988) (“[T]he preamble of a Jepson claim is impliedly admitted to be prior art.”).

As the Examiner points out, Barker discloses that tetrabutyl ammonium chloride, one of the water soluble phase transfer catalysts preferred by Appellants (FF 3, 4), was capable of improving the conversion rate in a aldol condensation reaction (FF 10, 11). As the Examiner also

points out, Halpern discloses that when water soluble quaternary ammonium salts like Barker's are used as phase transfer catalysts, they can be removed from the organic phase of the reaction milieu by washing with water (FF 17-20).

In view of these teachings, we agree with the Examiner that an ordinary artisan would have been prompted to include a water soluble phase transfer catalyst in a crossed-aldol condensation reaction, and remove the catalyst by water washing after performing the reaction.

Appellants do not dispute whether it would have been obvious to perform the cross aldol reaction using the specific aldehydes, propionaldehyde, etc., recited in claim 1. Rather, Appellants urge, an ordinary artisan would have been dissuaded from including a water soluble phase transfer catalyst in a crossed-aldol reaction in view of Kwok's disclosure that an MgO catalyst was "critical" to a crossed aldol reaction (App. Br. 11-13).

We are not persuaded. We acknowledge that Kwok's disclosure is directed to the use of the MgO catalyst, and that the MgO catalyst is therefore preferred by Kwok in the crossed-aldol reaction (FF 12, 15, 16). However, Kwok does not state that the MgO catalyst is critical to a crossed-aldol reaction, but rather that "[a] critical aspect *of the invention* is the use of hydrated MgO for catalyzing or promoting the condensation reaction" (Kwok, col. 4, ll. 58-59 (FF 16) (emphasis added)).

Thus, despite the preference for MgO, Kwok concedes that the crossed-aldol reaction proceeds "in the presence of alkali" (Kwok, col. 1, l. 64 (FF 14)). Accordingly, while Kwok may prefer its MgO catalyst, we do not agree that an ordinary artisan viewing Barker's disclosure of the

usefulness and potential advantages of the water soluble phase transfer catalyst tetrabutyl ammonium chloride would have been dissuaded by Kwok from using Barker's catalyst in crossed-aldol reactions.

Regarding the Judge patent, it might be true that page 4 lists only water insoluble phase transfer catalysts as useful in crossed aldol reactions. However, page 5 of Judge discloses that quaternary ammonium salts providing "excellent results" include those having alkyl groups from 1 to 20 carbon atoms (FF 27-28). Thus, in addition to disclosing quaternary ammonium salts with larger alkyl groups as phase transfer catalysts, Judge also discloses using, as phase transfer catalysts, quaternary ammonium salts with alkyl groups of the size present on Appellants' preferred catalysts (FF 28).

Regarding the asserted advantages of the process recited in claim 1, it may be true that Appellants have shown that certain water soluble phase transfer catalysts perform better than certain water insoluble phase transfer catalysts (FF 3). However, Appellants point to no evidence that an ordinary artisan would have considered this improvement unexpected in view of the prior art.

To the contrary, Barker discloses that one of the very water soluble phase transfer catalysts preferred by Appellants, tetra-butyl ammonium chloride, increased product conversion rate in an aldol condensation reaction (FF 10-11). Also, as noted above, Judge discloses that quaternary ammonium phase transfer catalysts with smaller alkyl groups produce "excellent results" in aldol condensation reactions (FF 27-28).

Moreover, as the Examiner points out, while Appellants have compared water soluble phase transfer catalysts to water insoluble ones, the

prior art actually discloses the use of water soluble catalysts. Given the prior art's disclosure of using water soluble phase transfer catalysts, Appellants have not adequately explained why the comparison presented in the Specification is a comparison to the closest prior art.

In sum, for the reasons discussed, we are not persuaded that the teachings in the cited references would have failed to prompt an ordinary artisan to practice the process recited in claim 1. As also discussed, we are not persuaded that the evidence of secondary considerations presented by Appellants is sufficient to outweigh the Examiner's obviousness case.

Accordingly, we affirm the Examiner's obviousness rejection of claim 1 over Barker in view of Kwok, Starks, Halpern, or Judge. As they were not argued separately, claims 3-9 and 12-28 fall with claim 1. *See* 37 C.F.R. § 41.37(c)(1)(vii).

TIME PERIOD

No time period for taking any subsequent action in connection with this appeal may be extended under 37 C.F.R. § 1.136(a).

AFFIRMED

cdc

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